NEW ANODE MATERIAL FOR RECHARGEABLE Li-ION CELLS

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INTRODUCTION

Carbon materials, such as graphite, cokes, pitch and PAN fibers, are being evaluated in lithium batteries as alternate anode materials'). Some degree of success has been achieved. However, the effort to look for other non-carbon anode materials which have larger Li capacity, higher rate capability, smaller first charge capacity loss and better mechanical stability during cycling is continuing. One possibility is Mg_2Si. From the Mg-Si binary **phase diagram, Mg_2Si is the** only compound formed between Mg and Si[2]. The compound Mg_2Si has a fluorite structure, which is face-centered cubic, with Si atoms at the cube corners and face centers, and Mg atoms occupying all the tetrahedral sites. This material was studied as an anode material at 400 ‘C using a molten salt electrolyte [3]. The proposed Li-Mg-Si ternary phase diagram[3] at 400 °C showed that intercalation of Li into the Mg_2Si will form the Mg and $Li_{13}Si_4$ phases. However, the electrochemical and thermodynamic properties of Li_xMg_2Si at room temperature was not studied. Thus, the feasibility of using Li_xMg_2Si as an alternate anode material at ambient temperature in organic electrolytes cells is not known. Recently, we have tried to intercalated Li into this material electrochemically at room temperature using an organic electrolyte. A voltage plateau at 260 mV (vs. Li) was observed, which is very attractive for using this material as an alternate anode. In this paper, the results of the electrochemical evaluation of Li_xMg_2Si material at ambient temperature will be presented.

EXPERIMENTAL

The anodes and cathodes were prepared by mixing powders with a binder until a uniform slurry was obtained. The slurry was spread on both sides of a nickel grid. The electrodes were then pressed between a set of stainless steel plates. Typically, electrodes had a coverage of 10 - **15** mg/cm^2, and were 10-15 mil thick. Electrochemical cells were constructed using these electrodes, lithium foil (Foote Mineral Corp.), porous polypropylene separators (Celgard no. 2400), and a electrolyte of 1.0M $LiPF_6$ in ethylene carbonate (EC) and dimethyl carbonate (DMC). Mg_2Si was firstly evaluated for its chemical stability or compatibility in the cell environment. The coulometric titration technique was applied to study the number of phases, phase composition, capacity and open circuit voltage (OCV vs. Li) of this material. To determine the general features of voltage as a function of composition, cells were charged and discharged using a constant current at ambient temperature. Experiments were conducted in a oxygen and moisture free dry box.

RESULTS AND DISCUSSION

The discharge curve shown in Figure 1 can be divided into three main regions based on the Li composition in Li_xMg_2Si. These are (1) $0<x<0.6$, (2) $0.6< x<1.0$ and (3) $x>1.0$. At x -0.6, the cell voltage was not stable and showed small periodic changes up to x - 1.0. However, further lithium intercalation gave a smooth voltage vs. composition curve. The voltage fluctuation in the discharge curve when Li intercalated into Mg_2Si may have something to do with the material's structure breakdown. The Li de-intercalation out of Li_xMg_2Si was also evaluated and the result is shown in Figure 1. **X-ray** diffraction (XRD) **analysis revealed that it is possible to** electrochemically **intercalated** about one Li per Mg_2Si and still kept the crystal structure. The XRD results did not show evidence for the existence of the Mg or $Li_{13}Si_4$ phases. The ambient temperature electrochemical intercalation of Li into Mg_2Si formed a single phase instead of multi-phases. Continued Li intercalation beyond the composition $Li_{1.0}Mg_2Si$ caused the material to become amorphous. It was determined that an additional mole of lithium could be intercalated into $Li_{1.0}Mg_2Si$. Preliminary results of the charge and discharge profiles and cycle life performance of cells containing a $LiCoO_2$ cathode and Mg_2Si anode are shown in Figure 2 and 3, respectively. Experimental results and observations indicated that the Mg_2Si electrode integrity was good and stable in the non-aqueous organic electrolyte.

SUMMARY

Experimental $Mg_2Si/LiCoO_2$ cells, containing an electrolyte of 1.0M $LiPF_6$ in EC + DMC, were fabricated and the cycle life performance of these cells was evaluated. These experimental cells have completed 70 cycles.

REFERENCES

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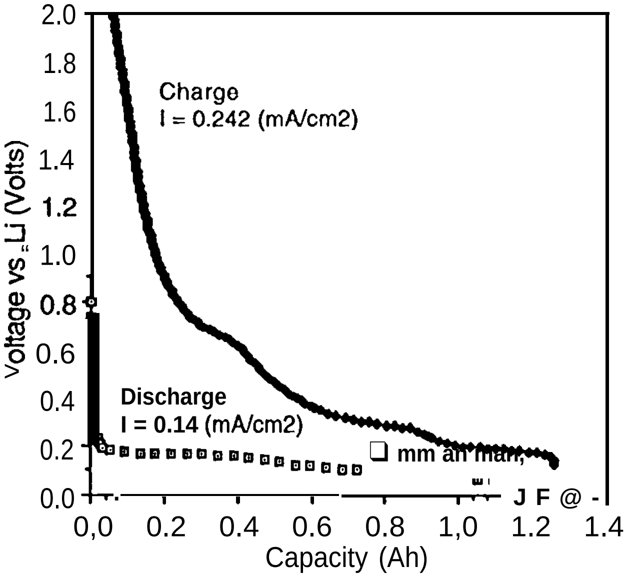

Figure 1. Charge and discharge profiles for the Li/ECDMC/Mg2Si cell.

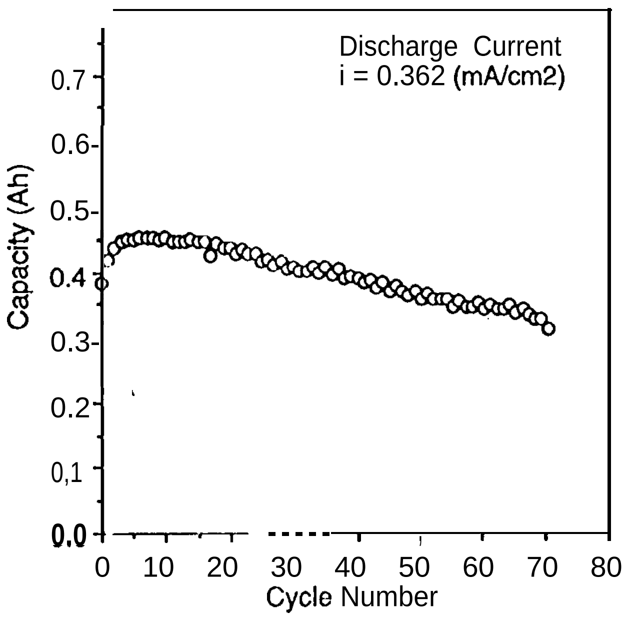

Figure 3. Cycle life performance for the Mg2Si/ECDMC/LiCoO2 cell.

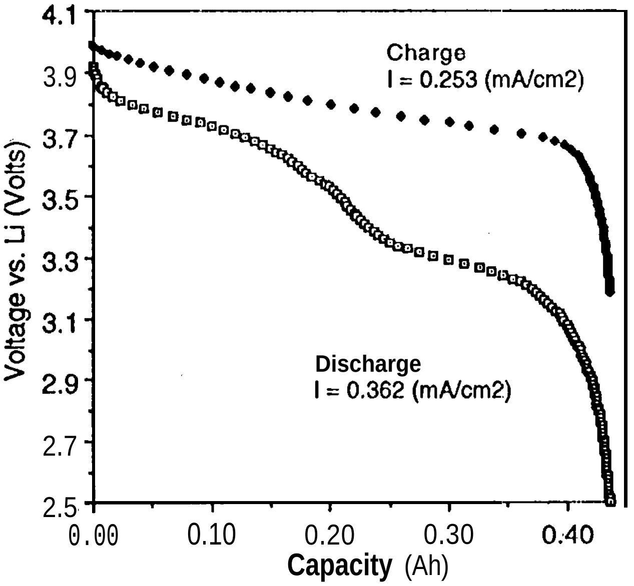

Figure 2. Charge and discharge profiles for the Mg2Si/ECDMC/LiCoO2 cell.